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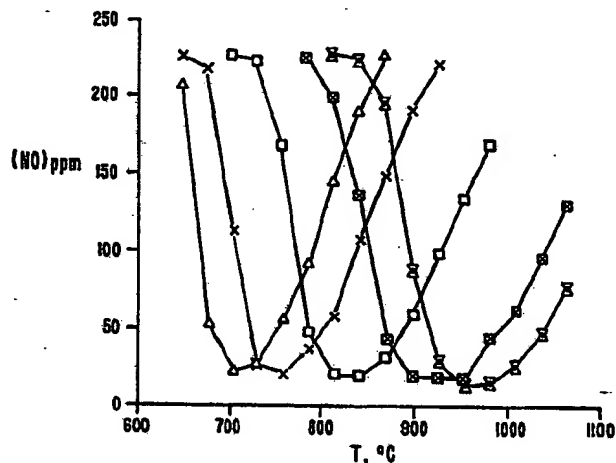
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(72) Inventors: HO, Loc ; 1014 N. Mayflower Street, Anaheim, CA 92801 (US). CHEN, Shih, L. ; 14 Alameda, Irvine, CA 92720 (US). SEEKER, William, Randall ; 711 Calle Brisa, San Clemente, CA 92672 (US). MALY, Peter, M. ; 25261 Tanoak Lane, El Toro, CA 92630 (US).

(54) Title: METHOD FOR CONTROLLING N_2O EMISSIONS AND FOR THE REDUCTION OF NO_x AND SO_x EMISSIONS IN COMBUSTION SYSTEMS

(57) Abstract

Methods for controlling N_2O emissions from stationary combustion systems having variable flue gas temperatures are disclosed. The N_2O emissions are controlled by the introduction of an N_2O control agent, such as an alkaline compound, into the effluent stream. In addition, the present invention discloses methods for controlling N_2O emissions from stationary combustion systems having variable flue gas temperatures while reducing NO_x emissions. Use of an NO_x reducing agent and an N_2O control agent, such as urea and monosodium glutamate, enlarges the temperature window for effective selective noncatalytic NO_x reduction while significantly eliminating N_2O emissions commonly experienced with urea injection. Further, the present invention discloses methods for controlling N_2O emissions from stationary combustion systems having variable flue gas temperatures while reducing SO_x emissions. Use of an NO_x reducing agent, an SO_x reducing agent, and an N_2O control agent, such as urea, an alkaline-earth metal compound, and an alkali-metal compound, respectively, enlarges the temperature window for effective NO_x and SO_x reductions while significantly eliminating N_2O emissions. Th figure is a graph illustrating how hydrogen mixed with ammonia shifts the temperature window of the selective non catalytic NO_x reduction process to lower temperatures.

Reduction of NO by H_2/NH_3 mixtures

$(NO)_0$ = 225 ppm, mass flow through reactor constant and corresponding to 0.1s at 1038 °C

⊠ for 0 - H_2/NH_3
⊞ for 0.125
□ for 0.50
× for 1.3
△ for 2.4

$(NH_3)_0 / (NO)_0$ = 2

METHOD FOR CONTROLLING N_2O EMISSIONS AND FOR THE REDUCTION OF NO_x AND SO_x EMISSIONS IN COMBUSION SYSTEMS

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BACKGROUND

1.   The Field of the Invention

The present invention is directed to an improved process for controlling N_2O emissions in combustion systems. More particularly, the present invention is directed to methods for controlling N_2O emissions by introducing a N_2O control agent into the effluent stream. In addition, the processes of the present invention significantly eliminate N_2O emissions while providing a wide temperature window for effective NO_x reduction by the use of an NO_x reducing agent. Further, the processes of the present invention significantly eliminate N_2O emissions while enhancing SO_x reduction by the use of an SO_x reducing agent.

2.   Technology Overview

The combustion of fossil fuels, industrial wastes, and municipal wastes produces significant amounts of NO_x and SO_x which ultimately participate in the formation of photochemical smog and acid rain. In addition to NO_x and SO_x, the combustion and incineration processes often produce another class of pollutants, namely, nitrous oxides (commonly referred to as "N_2O"). N_2O is a potential "greenhouse" gas and can also lead to degradation of the stratospheric ozone layer.

There exist many processes for the control of NO_x and SO_x emissions from power plants and incinerators. However, control of N_2O emissions from incinerators is a problem to which no presently available technology provides a fully satisfactory answer. While the amount of N_2O produced by burning fuel or waste can be minimized by managing the combustion process, waste or fuel typically contains substantial amounts of chemically bound nitrogen such that N_2O levels are usually unacceptably high, even with careful control of the combustion process. As a result, some form of post-combustion N_2O control technology must be used.

Wh reas the control of N_2O emissions is critical, the control of another class of pollutants - nitrogen oxides

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(commonly referred to as "NO_x") -is also important, and
current methods of reducing NO_x emissions result in the
formation of high N_2O emissions. While the amount of NO_x
produced by burning fuel or waste can be minimized by
5   managing the combustion process, as with N_2O emissions,
waste typically contains substantial amounts of chemically
bound nitrogen such that NO_x levels are usually unacceptably
high, even with careful control of the combustion process.
As a result, some form of post-combustion technology must
10   be used which not only controls N_2O emissions, but NO_x
emissions as well.

     Two types of post-combustion NO_x control technologies
are presently available, selective catalytic reduction
("SCR") and selective noncatalytic reduction ("SNCR").
15   Applications of SCR to incinerators are generally regarded
as nonfeasible because waste contains virtually all
possible trace impurities and these impurities can act as
catalyst poisons.

     As a result, SNCR processes have been adopted as the
20   best available NO_x control technology for incinerators. In
the usual SNCR process, a nitrogen-containing reducing
agent, normally either ammonia (NH_3) or urea (H_2NCONH_2), is
contacted with flue gas within a relatively narrow
temperature range. The optimum contacting temperature is
25   typically a factor of the reducing agent. A homogeneous
gas phase reaction occurs which reduces the NO_x in the flue
gas to molecular nitrogen (N_2) and water (H_2O).

     In most applications, the performance of the NO_x
reduction processes depends primarily on the available
30   reaction time, i.e., the length of time the flue gas spends
in the temperature range suitable for NO_x reduction by the
chosen reducing agent. For applications in which the
available reaction time is less than 0.2 seconds, NO_x
reductions in the 60% to 80% range are typically achieved.
35   For applicati ns in which the available reaction time is
greater than 0.2 seconds NO_x reducti ns in the 80% to 90%
range have c mmonly been achieved.

     The design of a modern incinerator provides the post-
flame gases with a residence time generally greater than

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1.0 seconds in th temperature range appropriate to NO_x
r duction processes. Hence, one might expect incinerators
to be a very favorable application for selective
noncatalytic NO_x reduction. Instead, however, NO_x reduction
in incinerators is typically only 40% or even less.

The poor performance of NO_x reduction processes on
incinerators is, in part, a result of the fact that the
temperature of the flue gas in incinerators is more highly
variable than it is in other combustion systems. Waste is
inherently a fuel with a highly variable BTU content. This
variability causes the temperature of the flue gases
downstream of the combustion zone to be nonhomogeneous in
space and to fluctuate in time.

If the temperature of the flue gas is a little too low
at the point where the reducing agent is injected, slight
or no NO_x reduction occurs. If the temperature is too high,
the nitrogen-containing reducing agent (NH_3 or H_2NCONH_2) has
some tendency to oxidize to produce NO_x, and the net
reduction of NO_x is poor or more NO_x may even be produced.
Because this range of temperatures ("temperature window")
for the NO_x reduction process is narrow, successful
application of the process is always critically dependent
on locating the reducing agent injection system at the
location at which the average temperature is optimum for
the process.

In any application, however, the temperature will be
nonhomogeneous, and process performance will be determined
by an average over a temperature range. Since this always
includes some temperatures which are too high and some
which are too low for good NO_x reduction, the practical
extent of NO_x control which the process can provide is
always significantly less than is achieved in laboratory
experiments.

Since the range of the NO_x r ducti n temperature window
increases with increasing reaction time, the longer
reaction time availabl in incinerators compensates, in
part, for this difficulty. However, there is an additi nal
problem: the optimum temperature for NO_x reduction may be
shifted. For example, as shown in Figure 1, (quoted from

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R.K. Lyon and J.E. Hardy, "Discovery and Development of the
Thermal DeNO$_x$ Process," 25 Ind. Eng. Chem. Fundam. 19
(1986); see also 21 Environmental Science and Technology,
232, (1987)) hydrogen (H$_2$) mixed with the ammonia shifts the
5   NO$_x$ reduction temperature window to lower temperatures. The
magnitude of the temperature shift increases as the amount
of H$_2$ is increased.

This shifting of the temperature window is a general
effect which occurs with other combustible materials,
10   including CO, natural gas, etc. Even though the
temperature window may be shifted, the size of the
temperature window is not enlarged to a significant degree
by the presence of other reducing agents in the combustion
effluent stream.

15   Whereas some reducing agents of NO$_x$ emissions, which
enlarge the useful temperature window for NO$_x$ reduction, are
known in the art, certain problems may result in their use.
While certain of the agents disclosed in this art reduce NO$_x$
emissions, the formation of N$_2$O emissions is also
20   experienced as discussed previously. Thus, benefits
incurred by some agents which can reduce NO$_x$ emissions is
negatively countered if those agents act to replace the NO$_x$
emissions with an alternate pollutant such as N$_2$O emissions.

The poor performance of N$_2$O and NO$_x$ reduction processes
25   also encounters a severe disadvantage due to the amount of
sulfur contained in the fuel to be incinerated. In most
instances, it is quite expensive and technically difficult
to remove the sulfur before burning the fuel. It has been
a common practice, therefore, to burn fuel which still
30   contains a portion of the native sulfur. Sulfur compounds,
including SO$_2$ and SO$_3$, are then produced during combustion
and must be removed from the effluent gas stream by
separate expensive technologies.

Combustion of a sulfur-containing fuel in a boiler,
35   furnace, or other burner typically produces combustion
effluents in which 98% to 99% of the sulfur exist in the
form of SO$_2$, and only 1% to 2% of the sulfur exists in the
form of SO$_3$. In processes where NO is reduced by injecting
ammonia into the gas stream, some of the ammonia or ammonia

precursors injected into th process will b left
unreacted. Under certain conditions, NH_3 will react with
the sulfur gases, _i.e._, as the combustion effluents cool,
the remaining NH_3 will react with SO_3 and water vapor
present in the effluent stream to form NH_4HSO_4 according to
reaction equation (1):

$$NH_3 + SO_3 + H_2O \rightarrow NH_4HSO_4$$

Unfortunately, NH_4HSO_4 is an extremely sticky and corrosive
liquid and is known to damage the equipment used in
combustion processes.

The temperature at which the formation of NH_4HSO_4
occurs is such that, in a typical boiler or furnace of the
type generally encountered in combustion processes,
formation of HN_4HSO_4 occurs within the air heater. Thus,
fouling, corrosion, and plugging of the air heater has been
commonly encountered. As a result, the nitric oxide
reduction technologies which use ammonia and ammonia
precursors have had severe limitations and their commercial
acceptance has been correspondingly limited. The operators
of boilers and furnaces are often extremely reluctant to
accept any technology which can cause fouling, plugging,
and corrosion such as that expected from NH_4HSO_4.

It will be noted that in the event that there are no
sulfur oxides in the effluent stream, the formation of
NH_4HSO_4 is not a problem. In many combustion applications,
however, there will be at least a small quantity of sulfur
oxides in the effluent stream and, thus, some SO_3. The
ammonia or ammonia precursors which are used in the nitric
oxide reduction reactions can then react with the SO_3
present to form NH_4HSO_4, which even in small quantities can
cause severe problems over time. Moreover, any SO_x
emissi ns which do n t form NH_4HSO_4, may react with water to
form a mist of sulfuric acid. This can also cause severe
problems over time.

From th foregoing, it is apparent that what is
currently needed in the art are methods for controlling N_2O

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emissions from stationary combustion systems having variable flue gas temperatures.

It would also be an improvement in the art if the present invention could provide methods for controlling N_2O emissions from stationary combustion systems while enlarging the useful temperature window and enhancing NO_x reduction.

It would be a further advancement in the art to provide methods for controlling N_2O emissions from stationary combustion systems which allows one to treat flue gas with ammonia or ammonia precursors to reduce NO_x emissions without producing an alternate class of pollutants such as N_2O.

It would be a still further advancement in the art to provide methods for controlling N_2O emissions from stationary combustion systems while enhancing SO_x reduction.

It would be a still further advancement in the art to provide methods for controlling N_2O emissions from stationary combustion systems while enlarging the useful temperature window for NO_x and SO_x reduction.

It would be an additional advancement in the art to provide methods for controlling N_2O emissions from stationary combustion systems which are economically feasible.

It would also be an additional advancement in the art to provide methods for controlling N_2O emissions from stationary combustion systems which perform effectively.

Such methods for controlling NO_x emissions from stationary combustion systems are disclosed and claimed herein.

BRIEF SUMMARY OF THE INVENTION

The present invention relates to methods for controlling N_2O emissions from stationary combustion systems. The present invention provides a process for controlling N_2O in emissions in effluent streams by introducing an N_2O controlling agent into the effluent stream within a certain temperature window. Preferably, the N_2O control agent is an alkaline compound. This process

significantly eliminates N_2O emissions from the combustion of th fu l.

Moreover, the present invention relates to methods for controlling N_2O emissions while reducing NO_x emissions from stationary combustion systems having variable flue gas temperatures. The present invention provides a process for controlling N_2O emissions while reducing NO_x emissions in effluent streams by introducing an NO_x reducing agent and an N_2O control agent into the effluent stream within a certain temperature window. Preferably, the NO_x reducing agent is ammonia or an ammonia precursor. The N_2O control agent is an alkaline compound, as defined at a later point. This process substantially reduces or eliminates the adverse effects of variable flue gas temperatures so as to reduce NO_x emissions while significantly eliminating N_2O emissions. The elimination of N_2O emissions is necessary because the injection of urea or ammonium/urea to effluents in stationary combustion systems to control NO_x emissions results in the production of higher N_2O emissions.

The present invention also provides a process for reducing SO_x emissions (which may or may not be present in effluent streams containing NO_x emissions) by introducing an SO_x reducing agent, and an N_2O control agent into the effluent stream within a certain temperature window. Preferably, the SO_x reducing agent is an alkaline earth-metal compound. The N_2O control agent is an alkaline compound. This process substantially reduces or eliminates the adverse effects of variable flue gas temperatures so as to reduce SO_x emissions while significantly eliminating N_2O emissions.

The present invention is further applicable to controlling N_2O emissions while reducing NO_x and SO_x emissions in effluent streams by introducing an NO_x and SO_x reducing ag nt, and an N_2O control agent into the effluent stream within a certain temperature window. Preferably, th NO_x reducing agent is ammonia or an ammonia precursor. The SO_x reducing agent is an alkaline-earth metal compound. The N_2O control agent is an alkaline compound, as described below. This process substantially reduces or eliminates

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the adverse effects of variable flue gas temperatures so as to reduce NO_x and SO_x emissions while significantly eliminating N_2O emissions.

5   BRIEF DESCRIPTION OF THE DRAWINGS

In order to better understand the manner in which the above-recited and other advantages and objects of the invention are obtained, a more particular description of the invention briefly described above will be rendered by

10   reference to a specific embodiment thereof which is illustrated in the appended drawings. Understanding that these drawings depict only a typical embodiment of the invention and are not therefore to be considered limiting of its scope, the invention will be described and explained

15   with additional specificity and detail through the use of the accompanying drawings in which:

Figure 1 is a graph illustrating how hydrogen mixed with ammonia shifts the temperature window of the selective noncatalytic NO_x reduction process to lower temperatures.

20   Figure 2 is a graph comparing NO_x reduction efficiency of urea and urea plus an amount of monosodium glutamate.

Figure 3 is a graph comparing N_2O emissions of urea and urea plus an amount of monosodium glutamate.

Figure 4 is a graph illustrating N_2O reduction

25   efficiency of aqueous Na_2CO_3.

Figure 5 is a graph illustrating SO_x reduction efficiency by the addition of alkali metals to limestone.

DETAILED DESCRIPTION OF THE PREFERRED EMBODIMENT

30   The present invention relates to methods for controlling N_2O emissions from stationary combustion systems such as power plant boilers, process furnaces, and incinerators. Additionally, the present invention relates to methods for controlling N_2O emissions while reducing NO_x

35   emissions from stationary combustion systems which have variable flue gas temperatures, the N_2O emissi ns being produced by the treatment of NO_x with ammonia or ammonia precursors. Moreover, the present invention relates to methods for controlling N_2O emissi ns while reducing SO_x

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emissions from stationary c mbustion systems. The present invention also relates to methods for controlling N_2O emissions while reducing NO_x and SO_x emissions from stationary combustion systems.

5   Specifically, the present invention is directed to a process for controlling N_2O in effluent streams having temperature variations and containing N_2O and excess oxygen. The process comprises the step of introducing an N_2O control agent into the effluent stream at a point where the
10   temperature of the effluent stream is in the range from about 1300°F to about 3000°F. Preferably, the process comprises the step of introducing an N_2O control agent into the effluent stream at a point where the temperature of the effluent stream is in the range from about 1800°F to about
15   2800°F.

The N_2O control agent of the process is an alkaline compound. The term "alkaline compounds" as used in this specification is meant to refer to compounds which comprise any of the univalent, mostly basic metals of Groups I and
20   II of the periodic table. These basic metals comprise lithium, sodium, potassium, rubidium, cesium, francium, magnesium and calcium. The alkaline compounds generally comprise alkaline-metal compounds of an organic acid, alkali-metal hydroxides, alkali-metal carbonates, alkaline-
25   earth metal compounds, and the like.

Some specific examples of alkaline compounds which fall within the scope of the present invention include sodium-based compounds (NaOH, Na_2SO_4, $NaHSO_4$, and Na_2CO_3), $Mg(CH_3COO)_2$ or $Ca(CH_3COO)_2$. Another example is monosodium
30   glutamate which is the monosodium salt of the naturally occurring L-form of glutamic acid. The compound is manufactured by the fermentation of carbohydrate sources. Monosodium glutamate is very soluble in water, and sparing soluble in alcohol. It must be rememb red, however, that
35   the present invention is not limited to th se specific examples, and that any compound fitting within and/or exhibiting th properties of alkaline compounds as broadly defined may also be included.

In one embodiment of the present invention, the N_2O control agent is an alkali-metal compound of an organic acid. Preferably, the N_2O control agent is an alkali-metal compound of an organic acid such as sodium glutamate,

5 potassium glutamate, sodium formate, potassium formate, sodium acetate, and potassium acetate. Nevertheless, it should be understood that, while the foregoing may be preferred under certain conditions, the present invention is not limited to the foregoing group of compounds.

10 In another embodiment of the present invention, the N_2O control agent is an alkali-metal hydroxide. Preferably, the N_2O control agent is an alkali-metal hydroxide selected from the group consisting essentially of LiOH, NaOH, and KOH. Nevertheless, it should be understood that the

15 present invention is not limited to the foregoing group of compounds.

In still another embodiment of the present invention, the N_2O control agent is an alkali-metal carbonate. Preferably, the N_2O control agent is an alkali-metal

20 carbonate selected from the group consisting essentially of lithium carbonate, lithium bicarbonate, sodium carbonate, sodium bicarbonate, potassium carbonate, and potassium bicarbonate. Nevertheless, it should be understood that the present invention is not limited to the foregoing group

25 of compounds.

In yet another embodiment of the present invention, the N_2O control agent is an alkaline-earth metal compound. Preferably, the N_2O control agent is an alkaline-earth metal compound selected from the group consisting essentially of

30 CaO, $CaCO_3$, $Ca(OH)_2$, $Ca(COOH)_2$, $Ca(CH_3COO)_2$, MgO, $MgCO_3$, $Mg(OH)_2$, $Mg(COOH)_2$, and $Mg(CH_3COO)_2$. Nevertheless, it should be understood that the present invention is not limited to the foregoing group of compounds.

An examination of the equivalent m le fraction of the

35 alkali and/or alkaline-earth metals in the N_2O control agent can also be determined. Preferably, the equivalent m le fraction of the alkali and/or alkaline-earth metals in the N_2O control agent is in the range from about 5 to about 2000 ppm by volume, in the effluent stream. Most preferably,

the N_2O control agent is in the range from about 50 ppm to
about 200 ppm by volume, in the effluent stream.

The amount of N_2O emissions that can be expected
following the use of the N_2O control agent are in the range
5   from about 0 ppm to about 50 ppm. Preferably, the limited
N_2O emissions which do occur in the range from about 0 ppm
to about 10 ppm.

As previously indicated, the present invention can be
successfully used to significantly suppress the formation
10   of N_2O in flue gases having a temperature in the range from
about 1300°F to about 2800°F at the point of contacting.
With regard to the N_2O control agent, the reaction time is
preferably greater than 0.1 seconds and more preferably
greater than 0.2 seconds. The temperature decay versus
15   time in the reaction zone of the present invention is
preferably less than about 600°F/second, and most
preferably less than about 200°F/second.

It should also be mentioned that the oxygen
concentration in the N_2O containing effluent streams shall
20   be in the range from about 0.1% to about 15% by volume of
oxygen to total flue gas. Preferably, the oxygen
concentration in the effluent streams is in the range from
about 2% to about 9%. It should be noted that the term
"controlling agent," as used throughout this application,
25   is meant to illustrate that there are limited amounts, if
any, of N_2O emissions. Controlling can mean that the N_2O
control agent suppresses the formation of N_2O emissions.
Controlling can also mean that the N_2O control agent
significantly eliminates N_2O emissions.
30   The present invention is also directed to a process
for controlling N_2O emissions while reducing NO_x in effluent
streams having temperature variations and containing NO_x and
excess oxygen. The process comprising the step of
intr ducing an NO_x reducing agent into an effluent stream at
35   a point where the temp rature f the effluent stream is in
the rang from about 1300°F to ab ut 2000°F, preferably in
the range from about 1600°F to about 1900°F.

The process also comprises the step of intr ducing an
N_2O control agent into the effluent stream at a point where

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the temperature of the effluent stream is in the range from
about 1300°F to about 3000°F, preferably in the range from
about 1800°F to about 2800°F. The N_2O control agent of this
process is similar to the N_2O control agent employed in the
5   process for controlling N_2O emissions.

The NO_x reducing agent of the process is ammonia and/or
an ammonia precursor, such as urea, ammonium sulfate,
ammonium carbonate, ammonium bicarbonate, ammonium formate,
ammonium acetate, and ammonium oxalate. Nevertheless, it
10   should be understood, however, that the NO_x reducing agent
is not limited to the foregoing group of compounds, but
would include other compounds capable of reducing NO_x
emissions.

Preferably, it has been found that by using a
15   combination of urea and alkaline compounds, over a range of
ratios, effective NO_x reduction can be obtained over a
significantly wider temperature range than by using
conventional noncatalytic NO_x reduction processes with
ammonia, urea, or a combination of ammonia and urea. In
20   this way, the adverse effects of variable flue gas
temperatures can be substantially reduced or eliminated and
the increased reduction of NO_x emissions can occur while the
control over the N_2O emissions is maintained.

Within the instant invention, the NO_x reducing agent
25   and the N_2O control agent may be injected into the flue gas
as a mist in a single step. Preferably, urea and alkaline
compound are injected into the flue gas in a single step.
The introduction of the compounds in a single step can be
differentiated from a step-wise or multi-stage process
30   wherein a plurality of treatment fluids are injected into
a combustion effluent stream at a plurality of temperature
zones. It can be understood that, not only does the single
stage use of urea and alkaline compound provide for
effective NO_x and SO_x reductions over a significantly wider
35   temperature range, but also that a singl stage step is
less complicated and less expensive.

Preparation of the solution containing urea and an
alkaline compound is accomplished in many different ways.
In one embodiment of the process for reducing NO_x, the N_2O

control agent and the NO_x reducing agent are mixed tog th r
and injected at the same location. Preferably, the urea
and an alkaline compound are premixed in a predetermined
ratio as a solution and injected in a single stage into the
flue gas. Alternatively, solutions containing the urea and
an alkaline compound, are impingently mixed in a single
stage into the flue gas.

Preferably, urea and alkaline compounds are typically
purchased in a solid form. Their use as a solution above,
therefore, requires that the solids be dissolved in some
type of a solvent. Examples of solvents include water and
alcohol; however, it should be understood that other
solvents, known to those skilled in the art, exist which
will dissolve urea and/or an alkaline compound.

In the preferred embodiment of the present invention,
an aqueous solution of urea is prepared, the concentration
of urea being in the range from about 2% to about 30%.
Thereafter, the aqueous urea and alkaline compound solution
is injected as a mist into the combustion effluent stream.

Alternatively, in another embodiment, the N_2O control
agent and the NO_x reducing agent are injected at separate
locations. The concentration of the urea solution is in
the range from about 2% to about 30%. The concentration of
the alkaline compound is in the range from about 0.25% to
about 7%.

The presence of an NO_x reducing agent in the flue gas
is in a predetermined stoichiometric ratio. The amount of
NO_x reducing agent, or specifically the urea, in the
effluent stream can also be determined by examining the
molar ratio of nitrogen in the NO_x reducing agent to the NO_x
in the effluent stream. Preferably, the molar ratio of
nitrogen in the NO_x reducing agent to the NO_x in the
effluent stream is in the range from about 0.5:1 to about
5:1. Most preferably, the molar ratio of nitrogen in the
NO_x reducing agent to the NO_x in the effluent stream is in
the range from about 1:1 to ab ut 3:1.

An examination of the equivalent mole fraction of the
alkali and/or alkaline-earth metals in the N_2O control agent
can also be determined. Preferably, the quivalent mole

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fraction of the alkali and/or alkaline-earth metals in the N_2O control agent is in the range from about 5 to about 2000 ppm by volume, in the effluent stream. Most preferably, the N_2O control agent is in the range from about 50 to about 5   200 ppm by volume, in the effluent stream.

It has also been found that the injection of an NO_x reducing agent and an N_2O control agent into a flue gas provides for the significant elimination of N_2O emissions in the flue gas. This is contrary to the injection of pure 10   urea or other urea combinations into flue gas which actually produces high N_2O emissions.

The amount of N_2O emissions that can be expected following the use of urea and an alkaline compound when compared to N_2O emission formed in the presence of urea 15   alone are in the range from about 0 ppm to about 20 ppm. Preferably, the limited N_2O emissions which do occur in the range from about 0 ppm to about 5 ppm.

As discussed above, it can be appreciated that the term "controlling agent," as used throughout this 20   application, is meant to illustrate that there are limited amounts, if any, of N_2O emissions. Controlling can mean that the N_2O control agent suppresses the formation of N_2O emissions. Controlling can also mean that the N_2O control agent significantly eliminates N_2O emissions.

25   As previously indicated, the present invention can be successfully used to reduce NO_x in flue gases having a temperature in the range from about 1300°F to about 2000°F at the point of contacting. With regard to the introduction of the N_2O control agent and the NO_x reducing 30   agent, the reaction time is preferably greater than 0.1 seconds and more preferably greater than 0.2 seconds. The temperature decay versus time in the reaction zone of the present invention is preferably less than about 600°F/second, and most preferably less than about 35   200°F/second.

The present invention can also be successfully used to significantly suppress the formation of N_2O in flu gases having a temperature in the range from ab ut 1300°F to about 2800°F at the point of contacting. With regard to

the N_2O control agent and the NO_x reducing agent, the reaction time is preferably greater than 0.1 seconds and more preferably greater than 0.2 seconds. The temperature decay versus time in the reaction zone of the present invention is preferably less than about 600°F/second, and most preferably less than about 200°F/second.

It should also be mentioned that the oxygen concentration in the N_2O and NO_x containing effluent streams shall be in the range from about 0.1% to about 15% by volume of oxygen to total flue gas. Preferably, the oxygen concentration in the effluent streams is in the range from about 2% to about 9%. The present invention is further applicable to controlling N_2O emissions while reducing SO_x emissions in effluent streams having temperature variations and containing SO_x and excess oxygen. It should be understood that in some instances, N_2O emissions occur in conjunction with SO_x emissions and not in the presence of NO_x emissions.

The process comprises the step of introducing an SO_x reducing agent into an effluent stream at a point where the temperature of the effluent stream is in the range from about 800°F to about 3000°F, preferably, in the range from about 1800°F to about 2800°F. The process further comprises the step of introducing an N_2O control agent into the effluent stream at a point where the temperature of the effluent stream is in the range from about 1300°F to about 3000°F, preferably, in the range from about 1800°F to about 2800°F.

The SO_x reducing agent of the process is an alkaline-earth metal compound. Preferably, the SO_x reducing agent is an alkaline-earth metal compound selected from the group consisting essentially of CaO, $CaCO_3$, $Ca(OH)_2$, $Ca(COOH)_2$, $Ca(CH_3COO)_2$, MgO, $MgCO_3$, $Mg(OH)_2$, $Mg(COOH)_2$, and $Mg(CH_3COO)_2$. Nevertheless, it should be understood that the present invention is not limited to this specific group of compounds.

Generally, the steps taken with regard to the SO_x reducing process are similar to the NO_x reducing process. Specifically, the N_2O control agent and the SO_x reducing

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agent are mixed together and injected at the same location
or the N_2O control agent and the SO_x reducing agent are
injected at separate locations. Also, the molar ratio of
the alkaline-earth metal in the SO_x reducing agent to the
5   SO_x in the effluent stream is in the range from about 0.5:1
to about 5:1. Additionally, the equivalent mole fraction
of the alkali metal in N_2O control agent is in the range
from about 10 to about 2000 ppm, by volume, in the effluent
stream. Further, the oxygen concentration in said effluent
10 streams is in the range from about 0.1% to about 15%,
preferably, in the range from about 2% to about 9%.

The present invention is also directed to a process
for controlling N_2O emissions while reducing NO_x and SO_x in
effluent streams having temperature variations and
15 containing NO_x, SO_x, and excess oxygen. The control of
pollutants in effluent streams naturally requires that in
some instances one would want to control the presence of
each of these three types of pollutants.

As discussed above, existing selective noncatalytic NO_x
20 reduction processes control NO_x emissions from stationary
combustion systems by injecting nitrogen-containing
reducing agents such as ammonia or ammonia precursors
(e.g., urea) either alone or in combination with a second
reducing agent, into the flue gases. The nitrogen-
25 containing reducing agent causes a homogeneous gas phase
reaction to occur which reduces NO_x to molecular nitrogen
and water. Unfortunately, selective noncatalytic NO_x
reduction processes that employ ammonia or urea are
ineffective since they lead to the unfavorable production
30 of N_2O emissions. Moreover, as discussed above, the use of
reducing agents such as ammonia or urea to treat flue gases
can lead to the undesirable formation of NH_4HSO_4 production
due to the presence of SO_x emissions. Thus, there may be
instances where one must control the presence f N_2O, NO_x,
35 and SO_x emissions.

The pr cess comprises the step of introducing an NO_x
reducing agent into an effluent stream at a point where the
temperature of the effluent stream is in the range from
about 1300°F to about 2000°F, preferably, in the range from

about 1600°F to about 1900°F . Th process also comprises
th step of introducing an SO_x reducing agent into an
effluent stream at a point where the temperature of the
effluent stream is in the range from about 800°F to about
3000°F, preferably, in the range from about 1800°F to about
2800°F. The process further comprises the step of
introducing an N_2O control agent into the effluent stream at
a point where the temperature of the effluent stream is in
the range from about 1300°F to about 3000°F, preferably, in
the range from about 1800°F to about 2800°F.

Generally, the steps taken with regard to the NO_x and
SO_x reducing process is similar to the NO_x and the SO_x
reducing processes discussed above. Specifically, the NO_x
reducing agent, SO_x reducing agent, and N_2O control agent of
this process is similar to the N_2O control agent employed in
the processes for reducing NO_x and SO_x emissions. In
addition, the molar ratio of nitrogen in the NO_x reducing
agent to the NOx in the effluent stream is in the range
from about 0.5:1 to about 5:1; the molar ratio of the
alkaline-earth metal in the SO_x reducing agent to the SO_x in
the effluent stream is in the range from about 0.5:1 to
about 5:1; the equivalent mole fraction of the alkali metal
in N_2O control agent is in the range from about 5 to about
2000 ppm, by volume, in the effluent stream; and the oxygen
concentration in said effluent streams is in the range from
about 0.1% to about 15%.

One distinction which must be made is in regard to the
introduction of the different agents in this process. In
one embodiment, the NO_x reducing agent, the N_2O control
agent, and the SO_x reducing agent are mixed together and
injected at the same location. In another embodiment, the
NO_x reducing agent, the N_2O control agent, and the SO_x
reducing agent are injected at different locations. In
still another embodiment, the NO_x reducing agent and the N_2O
control ag nt are injected at the same location but
separate from the injection l cation of the SO_x reducing
agent. In a further embodiment, the SO_x reducing agent and
the N_2O control agent are injected at the same location but
s parate from the injection location of the NO_x reducing

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agent.   In a still further embodiment, the SO_x reducing
agent and the NO_x reducing agent are injected at the same
location but separate from the injection location of the N_2O
control agent.

5   Experiments were conducted to determine the
performance of the N_2O control agent in eliminating N_2O
emissions.   Experiments were also conducted to determine
the performance of the N_2O control agent in eliminating N_2O
emissions while reducing NO_x emissions.   Experiments were
10   also conducted to determine the performance of the N_2O
control agent in eliminating N_2O emissions while reducing
SO_x emissions.

The following examples illustrate the suppression of
the formation of N_2O emissions achieved by the present
15   invention. The following examples further illustrate the
improved NO_x reduction achieved by the present invention
over a wide temperature range in addition to eliminating N_2O
emissions.   The following examples further illustrate the
improved SO_x reduction achieved by the present invention
20   over a wide temperature range in addition to eliminating N_2O
emissions.   These examples are intended to be, purely
exemplary of the use of the invention and should not be
viewed as limiting the scope of the present invention.

25   Example 1

In this example, the NO_x reduction efficiency (NO_f/NO_i)
of urea/monosodium glutamate and urea were compared over a
given temperature range. A 1×10^6 BTU/hour furnace fired
with natural gas at 3.5% excess oxygen was used to conduct
30   the experiment.   The initial NO_x concentration ("NO_i") in
the combustion effluents ranged from about 297 to about 303
ppm. The molar ratio of nitrogen in reducing agent to NO_i
was 1.5. The temperature decay versus time in the reaction
zone was approximately $400°F/second$.   The experimental
35   results are reproduced in Table 1 and shown graphically in
Figure 2.

Table 1

Chemical	NO$_i$ (ppm)	N/NO$_i$	Peak Temp. (°F)	NO$_f$/NO$_i$
Urea	298	1.50	1454	1.0
Urea	299	1.50	1614	0.86
Urea	303	1.50	1720	0.36
Urea	297	1.50	1847	0.32
Urea	298	1.50	2040	0.83
Urea + MSG	298	1.50	1454	0.5
Urea + MSG	299	1.50	1614	0.13
Urea + MSG	303	1.50	1720	0.12
Urea + MSG	297	1.50	1847	0.25
Urea + MSG	298	1.50	2040	0.89

Although the above NO$_x$ reduction efficiency with urea was higher at the optimum temperature than the NO$_x$ reduction efficiency with urea/monosodium glutamate, the temperature window for NO$_x$ reduction was much wider with urea/monosodium glutamate. The addition of monosodium glutamate significantly extended the reaction window to lower temperatures and enhanced the NO$_x$ reduction efficiencies with urea. Close to 90% NO$_x$ reduction was achieved at 1700°F with the addition of monosodium glutamate to urea.

Example 2

In this example, the N$_2$O reduction efficiency of urea/monosodium glutamate and urea were compared over a given temperature range. A 1 x 10^6 BTU/hour furnace fired with natural gas at 3.5% excess oxygen was used to conduct the experiment. The initial NO$_x$ concentration (NO$_i$) in the combustion effluents ranged fr m about 297 to about 303 ppm. The molar ratio of nitrog n in reducing agent to NO$_i$ was 1.5. The temp rature decay versus time in the reaction zon was approximately 400°F/second. The experimental

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results are r produced in Table 2 and shown graphically in Figure 3.

Table 2

Chemical	NO_i (ppm)	N/NO_i	Peak Temp. (°F)	N_2O (ppm)
Urea	298	1.50	1454	1
Urea	299	1.50	1614	4.5
Urea	303	1.50	1720	32
Urea	297	1.50	1847	40
Urea	298	1.50	2040	1
Urea + MSG	298	1.50	1454	7
Urea + MSG	299	1.50	1614	4
Urea + MSG	303	1.50	1720	3
Urea + MSG	297	1.50	1847	2
Urea + MSG	298	1.50	2040	0.5

The results of Example 2 confirm that the efficiency of N_2O reduction with urea depends strongly on the injection temperature, with the optimum near 1800°F. Use of urea/monosodium glutamate significantly eliminated the formation of N_2O emissions. Use of urea/monosodium glutamate also widens the temperature window for effective N_2O reduction. Using a 4:1 molar ratio of urea:monosodium glutamate, it is possible to achieve N_2O emissions in the range from about 0 ppm to about 8 ppm in a window between about 1450°F to about 2050°F.

Example 3

In this example, the NO_x reduction efficiency (NO_f/NO_i) of urea/sodium-containing compounds other than monosodium glutamate and urea were compared over a given temperature range. A 1 x 10⁶ BTU/hour furnace fired with natural gas at 3.5% excess oxygen was used to conduct the experim nt. The

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initial NO$_x$ concentration ("NO$_i$") in the combustion effluents ranged from about 298 to about 301 ppm. The ratio of reducing agent to NO$_i$ was 1.5. The temperature decay versus time in the reaction zone was approximately 400°F/second. The experimental results are reproduced in Table 3.

Table 3

Chemical	NO$_i$, ppm	N/NO$_i$	Peak Temp., °F	NO$_f$/NO$_i$
Urea	298	1.50	1454	1.0
Urea	299	1.50	1614	0.86
Urea	303	1.50	1720	0.36
Urea	297	1.50	1847	0.32
Urea	298	1.50	2040	0.83
Urea + NaOH	300	1.50	1850	0.27
Urea + NaOH	300	1.50	1740	0.23
Urea + Na$_2$CO$_3$	300	1.50	1850	0.27
Urea + Na$_2$CO$_3$	301	1.50	1840	0.23

Similar to Example 1, the temperature window for NO$_x$ reduction was much wider with urea/NaOH or Na$_2$CO$_3$ than with pure urea. The addition of these sodium-containing compounds significantly extended the reaction window to lower temperatures and enhanced the NO$_x$ reduction efficiencies with urea.

Example 4

In this example, the N$_2$O reduction efficiency of urea/sodium-containing compounds (other than monosodium glutamate) and urea were compared over a given temperature range. A 1 x 10^6 BTU/hour furnace fired with natural gas at 3.5% excess oxygen was used to conduct th experiment. The

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initial NO_x concentration (NO_i) in the combustion effluents

ranged from about 297 to about 303 ppm. The molar ratio of

nitrogen in reducing agent to NO_i was 1.5. The temperature

decay versus time in the reaction zone was approximately

5　400°F/second. The experimental results are reproduced in

Table 4.

Table 4

Chemical	NO_i (ppm)	N/NO_i	Peak Temp. (°F)	N_2O (ppm)
Urea	298	1.50	1454	1
Urea	299	1.50	1614	4.50
Urea	303	1.50	1720	32
Urea	297	1.50	1847	40
Urea	298	1.50	2040	1
Urea + NaOH	300	1.5	1850	0
Urea + Na_2CO_3	301	1.5	1850	0

The results of Example 4 confirm that the use of

simple sodium-containing compounds, such as NaOH and Na_2CO_3,

25　significantly eliminate the formation of N_2O emissions.

Using a 9:1 molar ratio of urea:sodium-containing

compounds, it is possible to eliminate N_2O emissions with

injections at 1850°F.

30　　　　　　　　　　　　　　## Example 5

An experiment was conducted similar to Example 2 with

the exception that a comparison is made between the N_2O

emission of urea/$Mg(CH_3COO)_2$ or $Ca(CH_3COO)_2$ and urea over a

giv n temperature range. The experimental results are

35　reproduced in Table 5.

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Table 5

Chemical	NO$_i$ (ppm)	N/NO$_i$	Peak Temp. (°F)	N$_2$O (ppm)
Urea	298	1.50	1454	1
Urea	299	1.50	1614	4.50
Urea	303	1.50	1720	32
Urea	297	1.50	1847	40
Urea	298	1.50	2040	1
Urea + Mg(CH$_3$COO)$_2$	300	1.5	1850	16
Urea+Ca(CH$_3$COO)$_2$	301	1.5	1850	12

Similar to Example 2, the results of Example 5 confirm that the use of urea/Mg(CH$_3$COO)$_2$ or Ca(CH$_3$COO)$_2$ significantly eliminated the formation of N$_2$O emissions. The additions of 1200 ppm (by volume) of Mg(CH$_3$COO)$_2$ or Ca(CH$_3$COO)$_2$ to urea resulted in N$_2$O reductions of about 60% and 70% respectively.

Example 6

An experiment was conducted similar to Example 2 except that a comparison is made between the N$_2$O emission of urea/Mg(CH$_3$COO)$_2$ and Na$_2$CO$_3$ and urea over a given temperature range. The experimental results are reproduced in Table 6.

Table 6

Chemical	NO$_i$ (ppm)	N/NO$_i$	Peak Temp. (°F)	N$_2$O (ppm)
Urea	298	1.50	1454	1
Urea	299	1.50	1614	4.50
Urea	303	1.50	1720	32
Urea	297	1.50	1847	40
Urea	298	1.50	2040	1
Urea+Mg+Na$_2$CO$_3$	300	1.5	1850	0

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Similar to Example 2, the results of Example 6 confirm that the use of urea/Mg(CH$_3$COO)$_2$ and Na$_2$CO$_3$ significantly eliminated the formation of N$_2$O emissions. The additions of about 20 ppm Na$_2$CO$_3$ to the mixture of urea and Mg(CH$_3$COO)$_2$

5   completely eliminated N$_2$O emissions.

Example 7

An experiment was conducted similar to Example 2 to compare the N$_2$O emission of urea/NaOH or Na$_2$CO$_3$ and urea over

10   a given temperature range. This example is similar to Example 4 with the exception that different concentrations of NaOH or Na$_2$CO$_3$ were employed. The experimental results are reproduced in Table 7.

15   Table 7

	NO$_i$ ppm	N/NOi	Temp°F	N$_2$O ppm
Urea	300	1.50	1740	40
Urea+NaOH(5-ppmNa)	300	1.50	1740	4
Urea+NaOH(20-ppmNa)	300	1.50	1740	1
Urea+Na$_2$CO$_3$ (20-ppm Na)	300	1.50	1740	0

20

25   Example 8

Under the same furnace conditions as in Example 1, experiments were also conducted with the promoter injected separately from the urea. With the aqueous urea solution injected at 1850°F, an aqueous solution of Na$_2$CO$_3$,

30   equivalent to 100 ppm sodium by volume, was inj cted at 2100, 1700, and 1550°F, separately from the urea. As shown in Table 8, all but the 1550°F injection case suppressed the formation of N$_2$O completely; 63% of N$_2$O was removed with the injection of Na$_2$CO$_3$ at 1550°F.

25

Table 8

	NO$_i$ ppm	N/NOi	Na$_2$CO$_3$ Temp°F	N$_2$O ppm
Urea @ 1850°F	300	1.50	NA	49
Urea @ 1850°F	300	1.50	2100	0
Urea @ 1850°F	300	1.50	1700	0
Urea @ 1850°F	300	1.50	1550	18

Example 9

Similar to Example 8, experiments were also carried out to study the control of N$_2$O emissions produced by sources other than urea, including combustion systems such as fluidized bed combustors or from other chemical processes. Under the same furnace conditions, N$_2$O gas was introduced at 1800°F to simulate the formation of N$_2$O from fluidized bed combustors. Aqueous sodium carbonate solution was injected at 2150°F for the removal of N$_2$O emissions. It can be seen from Figure 4 that the efficiency of N$_2$O reduction with sodium carbonate depended upon the initial N$_2$O concentration and the Na to N$_2$O molar ratio. Fifty percent reduction could be achieved at a sodium to N$_2$O ratio of 0.1.

The data presented in Figure 4 is also documented in Table 9.

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Table 9

SODIUM injected at 2150.°F
N$_2$O injected at 1800.°F

REDUCTION N$_2$O (PPM)	SODIUM/N$_2$O Molar Ratio	N$_2$O (%)
93	0.21	61.4
93	1.07	82.1
93	1.61	94.0
600	0.03	33.8
600	0.16	57.1
600	0.33	74.0
1350	0.03	35.0
1350	0.14	61.0
1350	0.16	65.0

Example 10

An experiment is conducted similar to Example 1 with the exception that the SO$_x$ reduction efficiency of limestone (CaCO$_3$) is determined. It is found that the addition of alkali-metal compounds to limestone significantly enhanced the SO$_x$ reduction efficiencies. Figure 5 presents these results.

From the foregoing, it will be appreciated that the present invention provides methods for controlling N$_2$O emissions from stationary combustion systems having variable flue gas temperatures.

It will also be appreciated that the present invention could provide methods for controlling N$_2$O emissions from stationary combustion systems while enlarging the useful temperature window and enhancing NO$_x$ reduction.

It will be further appreciated that the present invention provides methods for controlling N$_2$O emissions from stationary combustion systems which allows ne to treat flue gas with ammonia or ammonia precursors to reduce

NO_x emissions without producing an alternate class of pollutants such as N_2O.

It will be still further appreciated that the present invention provides methods for controlling N_2O emissions from stationary combustion systems while enhancing SO_x reduction. It will be still further appreciated that the present invention provides methods for controlling N_2O emissions from stationary combustion systems while enlarging the useful temperature window for NO_x and SO_x reduction.

It will be still further appreciated that the present invention provides methods for controlling N_2O emissions from stationary combustion systems which are economically feasible.

It will be still further appreciated that the present invention provides methods for controlling N_2O emissions from stationary combustion systems which perform effectively.

The invention may be embodied in other specific forms without departing from its spirit or essential characteristics. The described embodiments are to be considered in all respects only as illustrative and not restrictive. The scope of the invention is, therefore, indicated by the appended claims rather than by the foregoing description. All changes which come within the meaning and range of equivalency f the claims are to be embraced within their sc pe.

What is claimed is:

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1.   A process for controlling N_2O in effluent streams
having temperature variations and containing N_2O and excess
oxygen, comprising the step of introducing an N_2O control
agent into the effluent stream at a point where the
5   temperature of the effluent stream is in the range from
about 1300 °F to about 3000 °F, said N_2O control agent being
an alkaline compound of lithium, sodium, potassium,
rubidium, cesium, francium, magnesium or calcium.

10   2.   A process for controlling N_2O in effluent streams
as defined in Claim 1, having temperature variations and
containing N_2O and excess oxygen, comprising the step of
introducing an N_2O control agent into the effluent stream at
a point wherein the temperature of the effluent stream is
15   in the range from about 1800°F to about 2800°F.

3.   A process for controlling N_2O in effluent streams
having temperature variations and containing N_2O and excess
oxygen as in Claim 1, wherein the N_2O control agent is
20   introduced to the effluent stream to control N_2O produced by
the reaction of an NO_x reducing agent in the effluent
stream, said NO_x reducing agent being ammonia and/or an
ammonia precursor.

25   4.   A process for controlling N_2O in effluent streams
as defined in Claim 3, wherein the NO_x reducing agent is a
compound s lected from the gr up consisting f ammonia,
urea, ammonium oxalate, ammonium acetate, ammonium formate,
ammonium bicarbonate, ammonium carbonate, and ammonium
30   sulfate.

5.   A process for controlling N2O in effluent streams
as defined in Claim 1, wherein the N2O control agent is an
alkaline compound selected from the group consisting of
alkaline-metal compounds of an organic acid, alkali-metal
5   hydroxides, alkali-metal carbonates, and alkaline-earth
metal compounds.

6.   A process for controlling N_2O in effluent streams
as defined in Claim 1, wherein the N_2O control agent is a
10   compound selected from the group consisting of NaOH, Na_2SO_4,
$NaHSO_4$, Na_2CO_3, monosodium glutamate, potassium glutamate,
sodium formate, potassium formate, sodium acetate,
potassium acetate, LiOH, NaOH, KOH, lithium carbonate,
lithium bicarbonate, sodium carbonate, sodium bicarbonate,
15 .   potassium carbonate, potassium bicarbonate, CaO, $CaCO_3$,
$Ca(OH)_2$, $Ca(COOH)_2$, $Ca(CH_3COO)_2$, MgO, $MgCO_3$, $Mg(OH)_2$,
$Mg(COOH)_2$, and $Mg(CH_3COO)_2$.

7.   A process for controlling N_2O in effluent streams
20   as defined in Claim 1, wherein the molar ratio of the
alkali-metal or alkaline-earth metal in the control agent
to N_2O in the effluent stream is in the range from about
0.05:1 to about 1.5:1.

25   8.   A process for reducing NO_x while controlling the
emission of N_2O in effluent streams having temperature
variations and containing NO_x and xcess oxygen, comprising
the steps of:

(a)   introducing an NO_x reducing agent into an
30   effluent stream at a point where the temperature of

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the effluent stream is in the range from about 1300 °F to about 2000 °F, said NO_x reducing agent being ammonia and/or an ammonia precursor; and

(b) introducing an N_2O control agent into the effluent stream at a point where the temperature of the effluent stream is in the range from about 1300 °F to about 3000 °F, said N_2O control agent being an alkaline compound of lithium, sodium, potassium, rubidium, cesium, francium, magnesium or calcium.

9. A process for reducing NO_x while controlling the production of N_2O in effluent streams as defined in Claim 8, wherein the N_2O control agent is an alkaline compound selected from the group consisting of alkaline-metal compounds of an organic acid, alkali-metal hydroxides, alkali-metal carbonates, and alkaline-earth metal compounds.

10. A process for reducing NO_x while controlling the production of N_2O in effluent streams as defined in Claim 8, wherein the N_2O control agent is a compound selected from the group consisting of NaOH, Na_2SO_4, $NaHSO_4$, Na_2CO_3, monosodium glutamate, potassium glutamate, sodium formate, potassium formate, sodium acetate, potassium acetate, LiOH, NaOH, KOH, lithium carbonate, lithium bicarbonate, sodium carbonate, sodium bicarbonate, potassium carbonate, potassium bicarbonate, CaO, $CaCO_3$, $Ca(OH)_2$, $Ca(COOH)_2$, $Ca(CH_3COO)_2$, MgO, $MgCO_3$, $Mg(OH)_2$, $Mg(COOH)_2$, and $Mg(CH_3COO)_2$.

11. A process for reducing NO_x whil controlling the
production of N_2O in effluent streams as d fined in Claim 8,
wherein the NO_x reducing agent is a compound selected from
the group consisting of ammonia, urea, ammonium sulfate,
ammonium carbonate, ammonium bicarbonate, ammonium formate,
ammonium acetate, and ammonium oxalate.

12. A process for reducing NO_x while controlling the
production of N_2O in effluent streams as defined in Claim 8,
wherein the molar ratio of nitrogen in the NO_x reducing
agent to the NO_x in the effluent stream is in the range from
about 1:1 to about 3:1.

13. A process for reducing SO_x while controlling the
production of N_2O in effluent streams containing SO_x and
excess oxygen, comprising the steps of:

(a) introducing a SO_x reducing agent into an
effluent stream at a point where the temperature of
the effluent stream is in the range from about 800 °F
to about 3000 °F, said SO_x reducing agent being an
alkaline-earth metal compound; and

(b) introducing a N_2O control agent into the
effluent stream at a point where the temperature of
the effluent stream is in the range from about 1300 °F
to about 3000 °F, said N_2O control agent being an
alkaline compound of lithium, s dium, potassium,
rubidium, cesium, francium, magnesium or calcium.

14. A pr cess for reducing SO_x while controlling the
production of N_2O in effluent streams as defin d in

Claim 13, wherein the N_2O control agent is an alkaline compound selected from the group consisting of alkaline-metal compounds of an organic acid, alkali-metal hydroxides, alkali-metal carbonates, and alkaline-earth

5   metal compounds.

15.   A process for reducing SO_x while controlling the production of N_2O in effluent streams as defined in Claim 13, where the N_2O control agent is a compound selected

10   from the group consisting of NaOH, Na_2SO_4, $NaHSO_4$, Na_2CO_3, monosodium glutamate, potassium glutamate, sodium formate, potassium formate, sodium acetate, potassium acetate, LiOH, NaOH, KOH, lithium carbonate, lithium bicarbonate, sodium carbonate, sodium bicarbonate, potassium carbonate,

15   potassium bicarbonate, CaO, $CaCO_3$, $Ca(OH)_2$, $Ca(COOH)_2$, $Ca(CH_3COO)_2$, MgO, $MgCO_3$, $Mg(OH)_2$, $Mg(COOH)_2$, and $Mg(CH_3COO)_2$.

16.   A process for reducing SO_x while controlling the production of N_2O in effluent streams as defined in

20   Claim 13, wherein the molar ratio of the alkaline-earth metal in the SO_x reducing agent to the SO_x in the effluent stream is in the range from about 0.5:1 to about 5:1.

17.   A process for reducing NO_x and SO_x while

25   controlling the production of N_2O in effluent streams having temperature variations and containing NO_x, SO_x, and excess xygen, comprising the steps of:

            (a)   introducing an NO_x reducing agent into an
        effluent stream at a point where the temperature of

30   the effluent stream is in the range from about 1300 °F

to about 2000 °F, said NO$_x$ reducing agent being ammonia
and/or an ammonia precursor;

(b) introducing an SO$_x$ reducing agent into an
effluent stream at a point where the temperature of
the effluent stream is in the range from about 800 °F
to about 3000 °F, said SO$_x$ reducing agent being an
alkaline-earth metal compound; and

(c) introducing an N$_2$O control agent into the
effluent stream at a point where the temperature of
the effluent stream is in the range from about 1300 °F
to about 3000 °F, said N$_2$O control agent being an
alkaline compound selected from the group consisting
of alkaline-metal compounds of an organic acid,
alkali-metal hydroxides, alkali-metal carbonates, and
alkaline-earth metal compounds.

18. A process for reducing NO$_x$ and SO$_x$ while
controlling the production of N$_2$O in effluent streams as
defined in Claim 17, wherein the NO$_x$ reducing agent is a
compound selected from the group consisting of ammonia,
urea, ammonium sulfate, ammonium carbonate, ammonium
bicarbonate, ammonium formate, ammonium acetate, and
ammonium oxalate.

19. A process for reducing NO$_x$ and SO$_x$ while
contr lling the production of N$_2$O in effluent streams as
defined in Claim 17, wherein the molar ratio of nitr gen in
the NO$_x$ reducing agent to the NOx in the effluent stream is
in the range from about 0.5:1 to about 5:1.

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20. A proc ss for reducing NO_x and SO_x while controlling the production of N_2O in effluent streams as defined in Claim 17, wherein the molar ratio of the alkaline-earth metal in the SO_x reducing agent to the SO_x in the effluent stream is in the range from about 0.5:1 to about 5:1.

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Reduction of NO by H_2/NH_3 mixtures
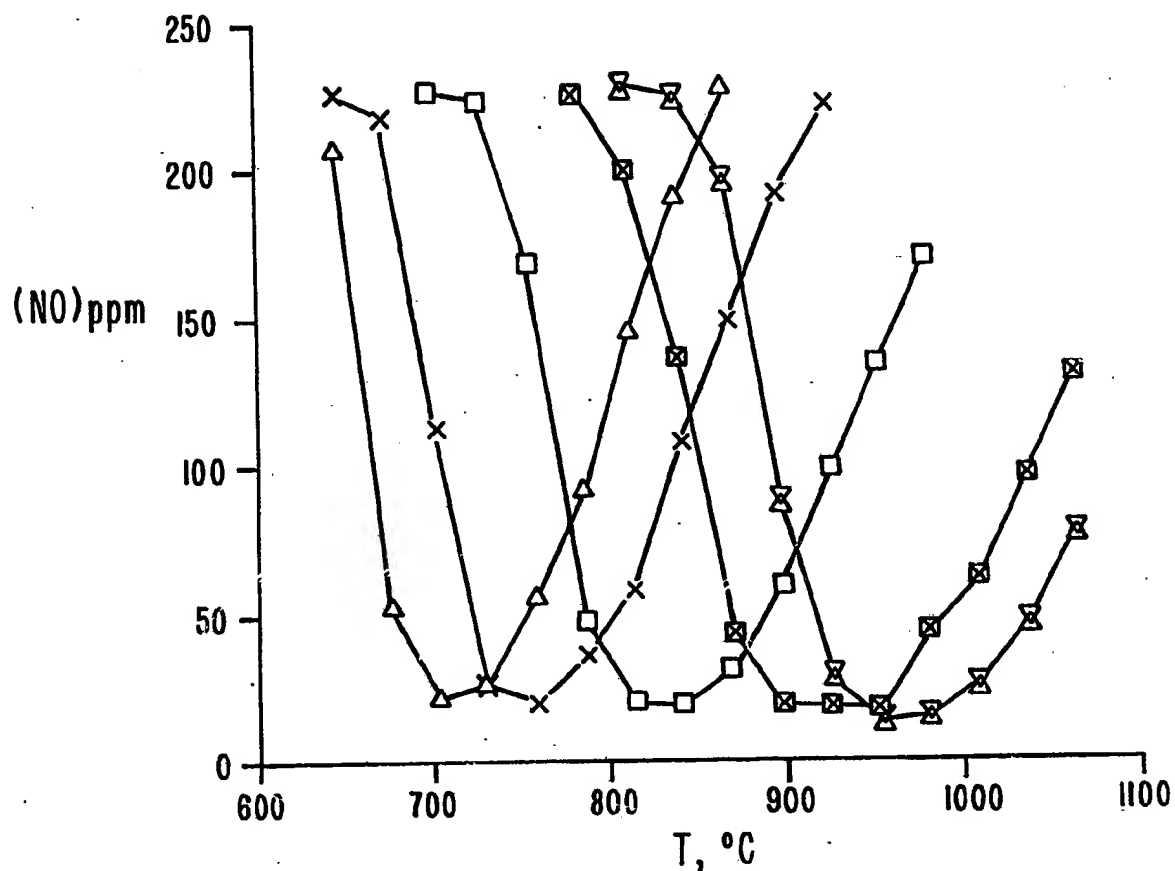
$(NO)_o = 225$ ppm, mass flow through reactor constant and corresponding to 0.1s at 1038 °C

⊠ for $0 = H_2/NH_3$

⊠ for 0.125

□ for 0.50
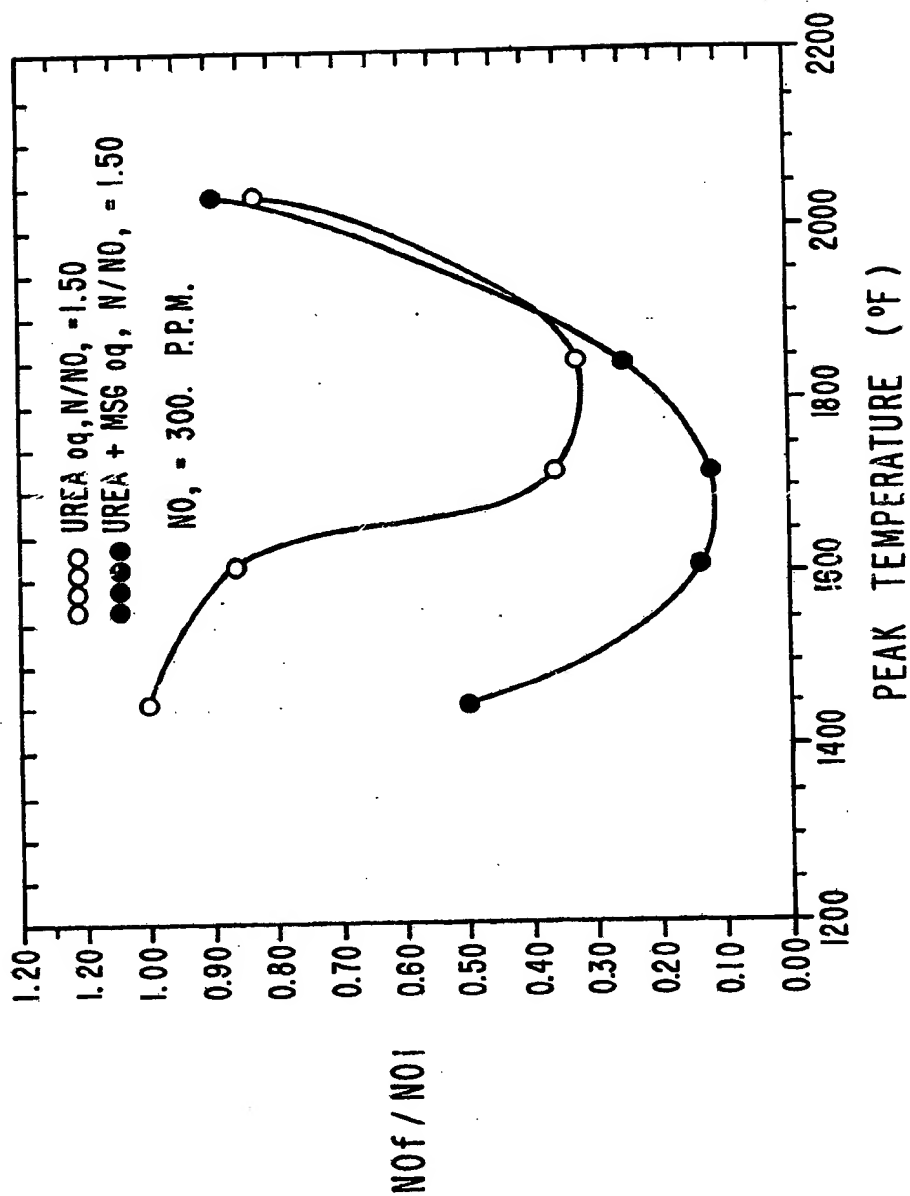
× for 1.3

△ for 2.4

$(NH_3)_0 / (NO)_0 = 2$
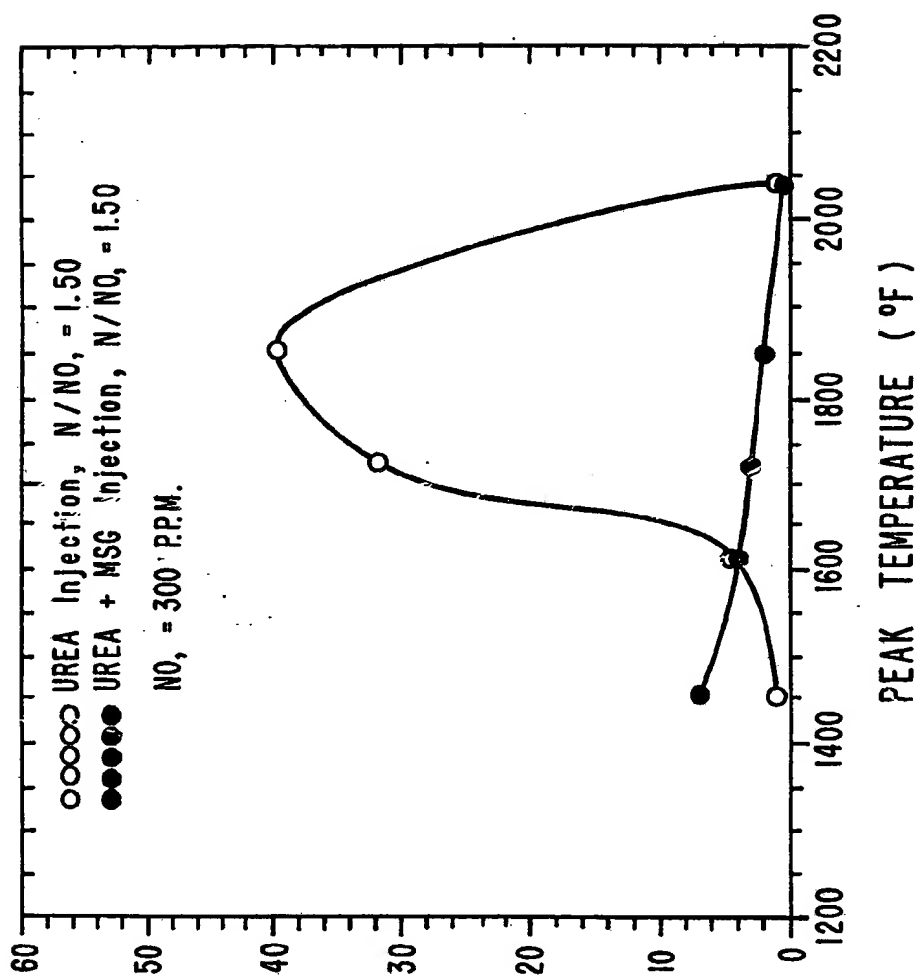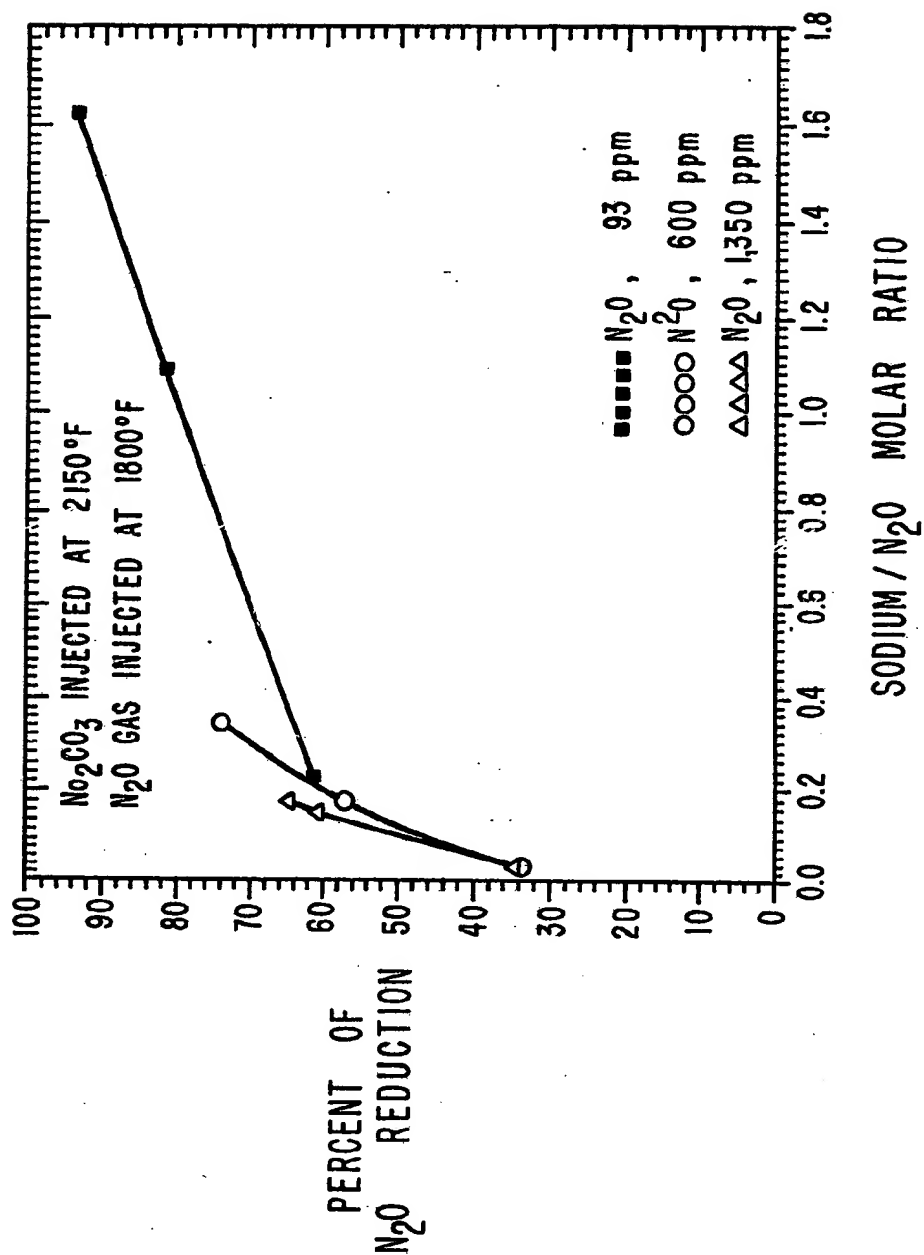
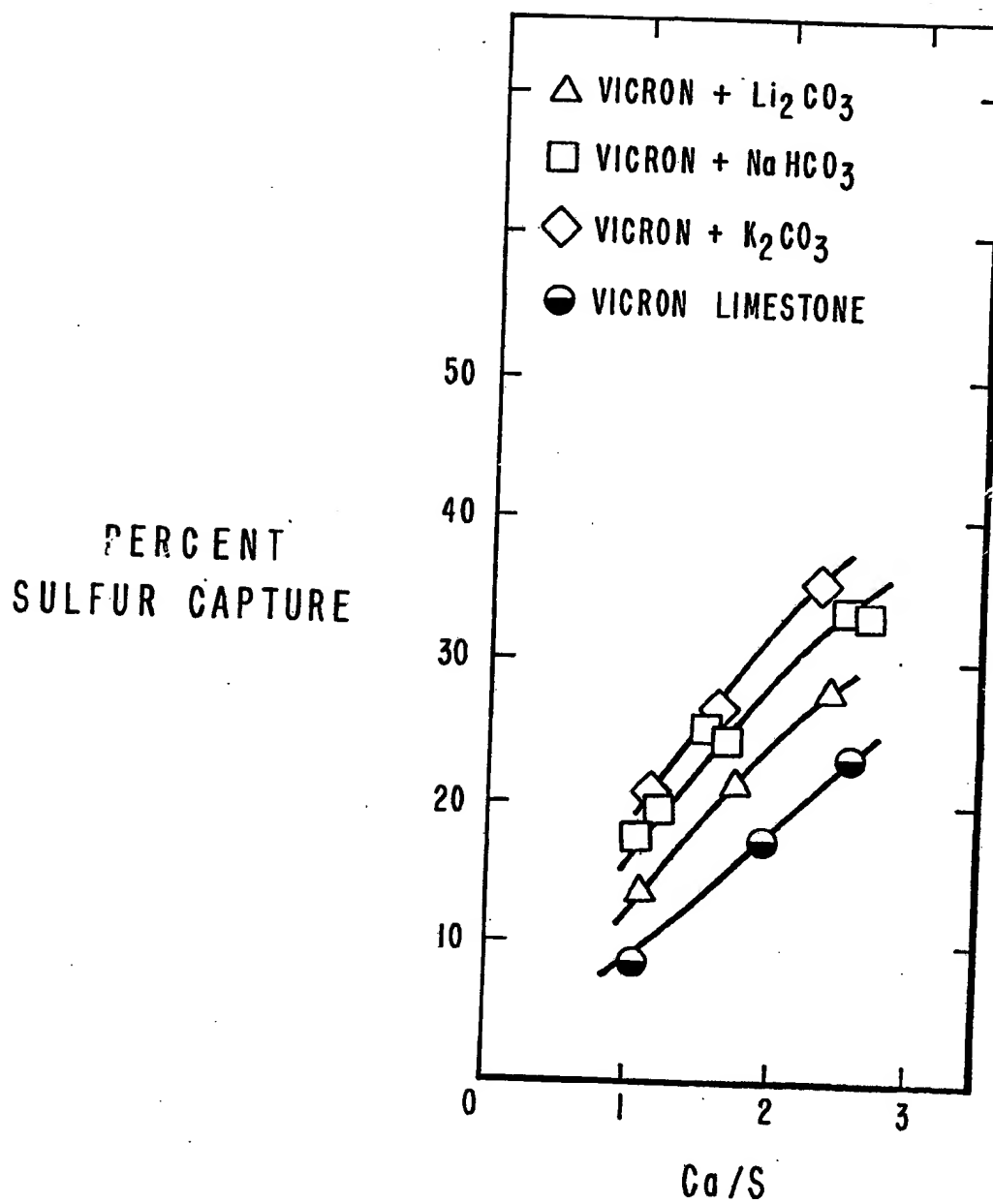## FIG. 1

FIG. 2

FIG. 3

FIG. 4

FIG. 5

INTERNATIONAL SEARCH REPORT

PCT/US93/02391

A.	CLASSIFICATION OF SUBJECT MATTER

IPC(5) :C01B 21/00
US CL :423/235,351

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

U.S. : 423/235, 351

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	US,A, 4,851,201 (HEAP ET AL) 25 JULY 1989	1-20
A	US,A, 4,861,567 (HEAP ET AL) 29 AUGUST 1989	1-20
A	US,A, 4,877,743 (WAUGH ET AL) 31 OCTOBER 1989	1-20
A	US,A, 5,048,432 (HOFMANN ET AL) 17 SEPTEMBER 1991	1-20

☐ Further documents are listed in the continuation of Box C.   ☐ See patent family annex.

*	Special categories of cited documents:	"T"	later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention
"A"	document defining the general state of the art which is not considered to be part of particular relevance		
"E"	earlier document published on or after the international filing date	"X"	document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone
"L"	document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)		
		"Y"	document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art
"O"	document referring to an oral disclosure, use, exhibition or other means		
"P"	document published prior to the international filing date but later than the priority date claimed	"&"	document member of the same patent family

Date of the actual completion of the international search	Date f mailing of the international search report
18 MAY 1993	01 JUL 1993

Name and mailing address of the ISA/US Commissioner of Patents and Trademarks Box PCT Washington, D.C. 20231	Authorized officer GREGORY A. HELLER
Facsimile No. NOT APPLICABLE	Telephone No. (703) 308-2533

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